RARE-EARTH, YTTRIUM AND SCANDIUM ABUNDANCES IN METEORITIC AND TERRESTRIAL MATTER—II

R. A. SCHMITT, R. H. SMITH and D. A. OLEHY

General Atomic Division of General Dynamics, John J. Hopkins Laboratory for Applied Science, San Diego 12, California

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Rare-earth, yttrium and scandium abundances in meteoritic and terrestrial matter—II*

R. A. Schmitt, R. H. Smith and D. A. Olehy *[handwritten: Repr. from]*

General Atomic Division of General Dynamics, John J. Hopkins Laboratory
for Pure and Applied Science, San Diego 12, California

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Abstract—Abundances and selected isotopic ratios of the 14 rare-earth elements (REE), yttrium, and scandium have been determined by neutron-activation analysis in 13 meteorites and 2 terrestrial specimens: 3 carbonaceous, 2 hypersthenic, and 1 bronzitic (troilite phase) chondrite; 2 calcium-rich and 2 nakhlitic achondrites; 1 mesosiderite; 2 pallasites; an Australian eclogite; and a Columbia Plateau basalt. This research is a continuation of recent REE work by Schmitt *et al.* (1963a). Absolute abundances of the REE and Sc in 2 Type I carbonaceous chondrites are about 33 per cent less compared with the Type II. Atomic ratios of $Y/10^6$ Si in carbonaceous chondrites remain approximately constant at 4·7 (Type I), 4·1 (Type II), and 4·8 (Type III), and ratios of the REE (La is representative) yielded $La/10^6$ Si at 0·36 (Type I), 0·53 (Type II), and 0·51 (Type III). The REE, Y, and Sc contents in the troilite-phase of a chondrite is ~0·3 of the content in the entire chondritic matrix. REE and Y abundances in Ca-rich achondrites and nakhlites are ~10 and ~5 times larger, respectively, than found in ordinary chondrites. No fractionation of the REE and Y distribution was observed in Ca-rich achondrites compared to chondritic REE and Y; fractionation in nakhlites is similar to terrestrial basalts. Fractionation of the REE and Y (Eu is enriched) in the mesosiderite Veramin and in two pallasites (Eu depleted in one pallasite) has been found. The observed fractionation of the REE and Y in Australian eclogite is opposite to fractionation in African eclogite, and fractionation in Columbia Plateau basalt was similar to Kilauea basalt.

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Introduction

In a recent paper by Schmitt and Smith (1962) and Schmitt *et al.* (1963a) (hereafter referred to as REE-I), the abundances and isotopic ratios have been determined for the 14 rare-earth elements (REE) plus Y and Sc by neutron-activation analysis in 19 meteorites and 3 terrestrial specimens. This paper is an extension of that work and reports the determination by neutron activation analysis of the absolute and relative abundances of the 14 REE, Y, and Sc in 13 meteorites and 2 terrestrial specimens. The neutron-activation procedure has been described in REE-I.

Results and Discussion

All the abundances obtained for the REE, Y, and Sc in chondritic and non-chondritic meteorites are summarized in Tables 1 and 2. The meteorites are classified according to Mason (1962a, 1962b). The normalized abundances of the REE, Y, and Sc are given in Table 3, where for comparative purposes, the average normalized REE, Y, and Sc abundances for 17 chondritic meteorites are given in the last column.

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Table 1. Absolute abundance of REE, Y and Sc in chondritic meteorites as determined by neutron-activation analysis[a]

Abundances in 10^{-6} g/g sample (ppm)

Isotope measured	Radio-nuclide	Carbonaceous chondrites			Hypersthenic chondrites		Bronzitic chondrite Richardton (Troilite Phase)
		Orgueil	Ivuna	Mighei	Modoc	Manbhoom	
La139	40-h La140	0·19 ± 0·01	0·19 ± 0·01	0·29 ± 0·01	0·34 ± 0·01	0·31 ± 0·01	0·74 ± 0·04[b]
Ce140	32-d Ce141	0·66 ± 0·01	0·60 ± 0·01	0·76 ± 0·02	1·42 ± 0·03	0·87 ± 0·01	—
Ce142	33-h Ce143	0·67 ± 0·01	0·57 ± 0·01	1·71 ± 0·01	1·59 ± 0·05	0·94 ± 0·01	—
Pr141	19-h Pr142	0·097 ± 0·001	0·090 ± 0·003	0·13 ± 0·01	0·13 ± 0·01	0·123 ± 0·005	0·058 ± 0·006
Nd146	11·1-d Nd147	0·44 ± 0·03	0·40 ± 0·02	0·61 ± 0·04	0·63 ± 0·02	0·65 ± 0·005	—
Sm152	47-h Sm153	0·131 ± 0·002	0·134 ± 0·003	0·20 ± 0·01	0·20 ± 0·01	0·244 ± 0·003	0·080 ± 0·004
Eu151	9.3-h Eu152 m	0·047 ± 0·002	0·053 ± 0·002	0·078 ± 0·001	0·076 ± 0·002	0·077 ± 0·002	—
Eu151	13-y Eu152	0·053 ± 0·002	0·057 ± 0·002	0·077 ± 0·003	0·084 ± 0·005	0·081 ± 0·001	—
Gd158	18-h Gd159	—	0·24 ± 0·04	0·38 ± 0·02	0·33 ± 0·01	0·26 ± 0·01	—
Tb159	72-d Tb160	0·022 ± 0·002	0·066 ± 0·003	0·047 ± 0·002	0·048 ± 0·001	0·059 ± 0·001	—
Dy164	2·3-h Dy164	0·22 ± 0·01	—	0·36 ± 0·07	0·30 ± 0·02	0·35 ± 0·02	—
Ho165	27-h Ho166	0·054 ± 0·001	0·058 ± 0·001	0·076 ± 0·002	0·084 ± 0·002	0·089 ± 0·002	0·034 ± 0·001
Er168	9·4-d Er169	0·151 ± 0·010	0·146 ± 0·005	0·24 ± 0·02	—	0·23 ± 0·02	—
Er170	7·5-h Er171	0·113 ± 0·009	0·151 ± 0·014	0·17 ± 0·01	0·23 ± 0·01	0·25 ± 0·01	—

Table 1 (cont.)

Abundances in 10^{-6} g/g sample (ppm)

Isotope measured	Radio-nuclide	Carbonaceous chondrites			Hypersthenic chondrites		Bronzitic Chondrite Richardton (Troilite phase)
		Orgueil	Ivuna	Mighei	Modoc	Manbhoom	
Tm^{169}	129-d Tm^{17}	0·022 ± 0·004	0·022 ± 0·002	0·030 ± 0·004	0·033 ± 0·002	0·036 ± 0·002	—
Yb^{168}	32-d Yb^{169}	0·14 ± 0·01	0·103 ± 0·002	0·17 ± 0·01	0·135 ± 0·004	0·21 ± 0·01	—
Yb^{174}	4·2-d Yb^{175}	0·17 ± 0·01	0·122 ± 0·003	0·17 ± 0·01	0·164 ± 0·003	0·18 ± 0·01	0·052 ± 0·007
Lu^{176}	6·8-d Lu^{177}	0·022 ± 0·001	0·024 ± 0·003	0·032 ± 0·005	0·036 ± 0·001	0·033 ± 0·001	0·010 ± 0·001
Y^{89}	64-h Y^{90}	1·44 ± 0·16	1·69 ± 0·19	1·77 ± 0·15	2·07 ± 0·08	1·97 ± 0·11	—
Sc^{45}	85-d Sc^{46}	5·3 ± 0·2	5·9 ± 0·3	8·6 ± 0·2	8·1 ± 0·2	8·8 ± 0·1	—

[a] Values for elemental abundances were calculated by assuming that terrestrial and meteoritic isotopic ratios were identical for all REE. This assumption has been verified for 5 REE isotopic ratios and particularly for the Yb^{168}/Yb^{174} ratio, which is strongly dependent on nucleosynthetic models. Errors are primarily one standard deviation, due to counting statistics. The chondritic meteorites have been classified according to MASON (1962a). See also MURTHY and SCHMITT (1963) for additional isotopic analyses of REE.

[b] The high La value in Richardton troilite may possibly be due to contamination because, on a normalized basis, only La is high by a factor of ~6 relative to the other REE.

Table 2. Absolute abundances of REE, Y, and Sc in achondritic, mesosideritic, and pallasitic meteorites[a]

| Isotope measured | Radionuclide | Achondrites (calcium-rich) | | Achondrites (Nakhlites) | | Mesosiderite | Pallasites | |
| | | | | | | Veramin (Silicate Phase) | Brenham (Olivine Phase) | Thiel Mts. (Olivine Phase) |
		Juvinas	Stannern	Nakhla	Lafayette			
La139	40-h La140	2·53 ± 0·05	4·89 ± 0·10	1·57 ± 0·05	1·76 ± 0·05	0·29 ± 0·01	—	0·12 ± 0·01[b]
Ce140	32-d Ce141	7·29 ± 0·15	12·6 ± 0·2	5·98 ± 0·06	5·50 ± 0·16	0·82 ± 0·02	—	—
Ce142	33-h Ce143	7·20 ± 0·15	13·2 ± 0·2	6·40 ± 0·19	5·46 ± 0·13	0·91 ± 0·03	≤0·17 ± 0·01[c]	0·28 ± 0·03
Pr141	19-h Pr142	0·98 ± 0·03	1·96 ± 0·04	0·67 ± 0·03	0·80 ± 0·03	0·114 ± 0·005	0·032 ± 0·002	0·022 ± 0·001[d]
Nd146	11-1-d Nd147	5·01 ± 0·12	10·0 ± 0·2	3·18 ± 0·2	3·35 ± 0·06	0·57 ± 0·07	0·16 ± 0·01	0·12 ± 0·01
Sm152	47-h Sm153	1·74	3·17 ± 0·06	0·73 ± 0·02	0·85 ± 0·02	0·23 ± 0·01	0·112 ± 0·002	0·022 ± 0·001
Eu151	9-3-h Eu152m	0·60 ± 0·02	0·86 ± 0·02	0·20 ± 0·01	0·24 ± 0·01	0·113 ± 0·002	0·018 ± 0·001	0·0031 ± 0·0001
Eu151	13-y Eu152	0·64 ± 0·02	0·80 ± 0·03	0·19 ± 0·01	0·23 ± 0·01	0·122 ± 0·005	0·017 ± 0·001	—
Gd158	18-h Gd159	2·55 ± 0·05	4·47 ± 0·09	0·94 ± 0·03	0·92 ± 0·04	0·44 ± 0·02	—	0·020 ± 0·003
Tb159	72-d Tb160	0·40 ± 0·01	0·71 ± 0·02	0·109 ± 0·002	0·120 ± 0·003	0·054 ± 0·004	0·038 ± 0·002	—
Dy164	2-3-h Dy165	3·03 ± 0·10	4·89 ± 0·14	—	0·89 ± 0·14	0·37 ± 0·01	—	0·016 ± 0·003
Ho165	27-h Ho166	0·59 ± 0·01	1·07 ± 0·03	0·140 ± 0·002	0·146 ± 0·004	0·097 ± 0·002	0·051 ± 0·002	0·0034 ± 0·0002
Er168	9-4-d Er169	1·70 ± 0·13	2·98 ± 0·15	0·36 ± 0·02	0·47 ± 0·03	0·27 ± 0·03	0·097 ± 0·006	—
Er170	7-5-h Er171	1·85 ± 0·03	3·12 ± 0·05	0·32 ± 0·01	0·40 ± 0·01	0·25 ± 0·01	0·091 ± 0·004	—

Abundances in 10⁻⁶ g/g sample (ppm)

Table 2 (cont.)

Abundances in 10^{-6} g/g sample (ppm)

Isotope measured	Radionuclide	Achondrites (calcium-rich)		Achondrites (Nakhlites)		Mesosiderite	Pallasites	
		Juvinas	Stannern	Nakhla	Lafayette	Veramin (Silicate phase)	Brenham (Olivine phase)	Thiel Mts. (Olivine phase)
Tm^{169}	129-d Tm^{170}	0·28 ± 0·01	0·47 ± 0·02	0·047 ± 0·003	0·057 ± 0·006	0·041 ± 0·005	0·0096 ± 0·0009	0·0010 ± 0·0002
Yb^{168}	32-d Yb^{169}	1·37 ± 0·04	2·38 ± 0·06	0·21 ± 0·01	0·28 ± 0·02	0·19 ± 0·01	—	—
Yb^{174}	4·2-d Yb^{175}	1·37 ± 0·04	2·36 ± 0·08	0·24 ± 0·01	0·36 ± 0·01	0·23 ± 0·01	0·043 ± 0·001	0·0086 ± 0·0005
Lu^{176}	6·8-d Lu^{177}	0·231 ± 0·004	0·38 ± 0·01	0·044 ± 0·001	0·051 ± 0·002	0·044 ± 0·002	0·0059 ± 0·0004	0·0017 ± 0·0002
Y^{89}	64-h Y^{90}	17·1 ± 0·3	28 ± 0·5	3·17 ± 0·15	4·4 ± 0·2	2·47 ± 0·17	1·57 ± 0·19	—
Sc^{45}	85-d Sc^{46}	28 ± 0·4	31 ± 0·6	54 ± 1·1	78 ± 1·5	16·2 ± 0·3	0·79 ± 0·03	1·15 ± 0·02

[a] Values for elemental abundances were calculated by assuming that terrestrial and meteoritic isotopic ratios were identical for all REE. This assumption has been verified for 5 REE isotopic ratios and particularly for the Yb^{168}/Yb^{174} ratio, which is strongly dependent on nucleosynthetic models. Errors are primarily one standard deviation, due to counting statistics. The chondritic meteorites have been classified according to MASON (1962a). See also MURTHY and SCHMITT (1963) for additional isotopic analyses of REE.

[b] The La value for another olivine portion in Thiel Mountains was 0·022 ± 0·001 ppm.

[c] The upper limit to Ce was calculated because of the presence of Ce^{144} gamma rays that were possibly due to neutron-induced fission of uranium in olivine.

[d] The value given for Pr was obtained from the first neutron-activation analysis of another olivine portion of Thiel Mountains. In the two Thiel Mountains analyses the La/Pr ratios were 5·5 and 1·8, respectively. The source of the discrepancy is unknown, but the value given for Pr agrees well with the other REE when normalized to that of the chondrites.

Table 3. Normalized abundances of REE, Y and Sc in chondritic, achondritic, mesosideritic and pallasitic meteorites

Element	Chondrites						Achondrites (calcium-rich)		Achondrites (Nakhlites)		Meso-siderite	Pallasites (Olivine phase)		Average in 17 chondrites[a]
	Orgueil (Carb.)	Ivuna (Carb.)	Mighei (Carb.)	Modoc (Hyp.)	Man-bhoom (Hyp.)	Richardton (Bron Troilite)	Juvinas	Stannern	Nakhla	Lafayette	Veramin	Brenham	Thiel Mts.	
La	1·00	1·00	1·00	1·00	1·00	6·1[b]	1·00	1·00	1·00	1·00	1·00	(b)	1·00	1·00
Ce	3·50	3·08	2·62	4·40	2·81	—	2·86	2·64	3·94	3·12	2·98	≤ 1·00	2·33	2·80 ± 0·23[c]
Pr	0·51	0·47	0·45	0·38	0·40	0·48	0·39	0·40	0·43	0·45	0·39	0·19	0·18	0·41 ± 0·005
Nd	2·31	2·10	2·10	1·85	2·10	—	1·98	2·05	2·02	1·90	1·97	0·91	1·01	1·88 ± 0·16
Sm	0·69	0·71	0·69	0·59	0·78	0·66	0·69	0·65	0·47	0·48	0·79	0·66	0·18	0·66 ± 0·05
Eu	0·26	0·29	0·27	0·24	0·25	—	0·25	0·176	0·124	0·134	0·40	0·13	0·026	0·24 ± 0·03
Gd	—	1·26	1·31	0·97	0·84	—	1·01	0·92	0·60	0·52	1·52	—	0·17	0·97 ± 0·11[d]
Tb	0·12	0·35	0·16	0·14	0·19	—	0·158	0·145	0·069	0·068	0·19	0·22	—	0·16 ± 0·02
Dy	1·16	—	1·24	0·88	1·13	—	1·20	1·00	—	0·51	1·28	—	0·13	0·99 ± 0·13
Ho	0·28	0·31	0·26	0·25	0·29	0·28	0·23	0·22	0·089	0·083	0·34	0·30	0·027	0·24 ± 0·03
Er	0·68	0·79	0·71	0·68	0·81	—	0·70	0·63	0·216	0·247	0·90	0·55	—	0·69 ± 0·06
Tm	0·116	0·116	0·103	0·097	0·116	—	0·111	0·096	0·030	0·032	0·141	0·057	0·0083	0·103 ± 0·008[e]
Yb	0·82	0·60	0·52	0·45	0·63	0·43	0·54	0·48	0·143	0·182	0·73	0·25	0·072	0·56 ± 0·05[f]
Lu	0·116	0·126	0·11	0·106	0·11	0·082	0·091	0·078	0·028	0·028	0·152	0·035	0·014	0·105 ± 0·012
Y	7·6	8·9	6·1	6·1	6·4	—	6·8	5·7	2·02	2·50	8·5	9·2	—	6·5 ± 0·5[g]
Sc	28	31	30	24	28	—	11·0	6·2	34	44	56	4·7	9·6	27 ± 3[h]

[a] Average values include 12 chondrites given in Table 3 of SCHMITT et al. (1963a) and 5 chondrites of this Table. Average abundance of La = 0·30 ppm in 17 chondrites.
[b] Abundances normalized to Sm at 0·66.
[c] Abundances of Allegan, Richardton, Holbrook, Modoc, and Orgueil excluded.
[d] Abundance of Ivuna excluded.
[e] Abundance of Forest City excluded.
[f] Abundance of Orgueil excluded.
[g] Abundance of Ivuna excluded; average of 11 chondrites given.
[h] Abundance of Holbrook excluded; average of 11 chondrites given.

It may be remarked parenthetically that the contribution to Ce^{141} and Ce^{143} from the neutron-induced fission of the uranium and thorium present in these chondrites is only about 1–2 per cent and therefore negligible. The constancy of the meteoritic Ce^{141}/Ce^{143} ratio relative to a pure Ce standard and the absence of enhanced Nd abundance via the 11·1-d Nd^{147} radiostope (also a possible fission product) rules out significant neutron-fission contribution to the REE abundances.

General abundances in chondritic meteorites

1. Within experimental error, most of the normalized REE and Sc abundances in Orgueil and Ivuna agree with the normalized average in 17 chondrites. On an absolute basis, the concentrations of the REE and Sc in Orgueil and Ivuna are rather low, i.e. the abundance of La, a representative REE, is 0·19 ppm, whereas the average La abundance in 17 chondrites is 0·30 ppm.

An atomic ratio of $La/10^6$ Si is calculated at 0·36 $\pm$ 0·01 for Type I Orgueil and Ivuna, and 0·53 $\pm$ 0·07 is found in Type II carbonaceous chondrites Mighei and Murray (see REE-I and Table 1 of this paper). $La/10^6$ Si values of 0·51 $\pm$ 0·08, 0·38 $\pm$ 0·07, 0·38 $\pm$ 0·03, and 0·24 $\pm$ 0·04 were obtained for pigeonitic, hypersthenic, bronzitic, and enstatitic chondrites, respectively. These atomic ratios indicate that the enstatitic chondrites deviate significantly from the other four chondritic categories, i.e. the contents of the trace REE and Sc appear to be markedly depleted relative to Si in enstatitic chondrites. On the basis of weight (H_2O and C excluded), the abundances of the REE and Sc in Type I carbonaceous and enstatitic chondrites agree well (these are discussed in a later section, as are the abundances of Y).

The low atomic ratio of the trace element La at 0·36 for Type I carbonaceous chondrites compared with higher values in other carbonaceous classes may result from some inhomogeneity of chondritic matter during the initial accretion of primitive matter (WOOD, 1962) or as a result of subsequent physiochemical processes. Since abundances in the percentile range of such elements as Al, S, Na, and Ca fluctuate appreciably in the various carbonaceous classes (WIIK, 1956), variations in trace-element abundances within the carbonaceous chondritic classes should not be unexpected.

2. The normalized abundances of Ce and Yb appear to be enhanced in Orgueil relative to the average in 17 chondrites. With Tm and Lu, which are adjacent to Yb, apparently unaffected in normalized abundances, no account can be offered for the apparent discrepancy of Yb. One measure of the meteoritic oxidation history may be found in the comparisons of the Ce/La ratios. A high Ce/La ratio of 3·50 in Orgueil may be indicative of an above-average oxidation environment during mineral formation compared to the environment of Types I and II carbonaceous chondrites Ivuna, Murray, and Mighei, which have lower Ce/La ratios of 3·08, 2·69, and 2·55, respectively (see REE-1). The element Ce, upon oxidation to the +4 state, may crystallize in lattices different from the common trivalent REE minerals. Since such processes are ordinarily restricted to highly mobile and diffusive conditions common to the liquid or semiliquid state at high temperatures, it is highly probable that any organisms, if present, would be destroyed during this particular phase of chondritic history. (The possible existence of fossilized organisms in Type I carbonaceous chondrites has been discussed at some length in "Life Forms in Meteorites," 1962.)

The high Ce/La ratio in Orgueil may be interpreted in the light of the recent discussion by Dufresne and Anders (1962) on the chemical evolution of the carbonaceous chondrites. Evidence has been presented which suggest that these carbonaceous meteorites and the associated minerals have been exposed to the chemical action of water. If the REE had precipitated in separate minerals, such as carbonates, and had been exposed to an oxidizing alkaline medium for oxidation of Ce^{+3} to Ce^{+4} ($E° = -1.61$), percolation of water through such a mineral system would have preferentially dissolved the trivalent REE, e.g. the solubility product of $Ce(OH)_4$ at 25°C is $\sim 10^{-48}$, whereas the $REE(OH)_3$ solubility products vary uniformly from $\sim 10^{-19}$ to $\sim 10^{-24}$ for $La(OH)_3$ through $Lu(OH)_3$. (Dolomite, a carbonate, has been found in both Orgueil and Ivuna.) The observed $La/10^6$ Si atomic ratio of 0.36 could be explained if $\sim 1/3$ of the trivalent REE initially present (in amounts corresponding to the average of 0.53 ± 0.07 $La/10^6$ Si in Type II Mighei and Murray) were dissolved. Retention of all Ce^{+4} in the REE mineral upon water erosion would yield a Ce/La ration of 3.5, which agrees with the observed value of 3.50. If the REE are scattered homogeneously through the common silicate minerals, one third of these silicate minerals must necessarily be dissolved.

Dissolution of small REE minerals with large surface-to-volume ratios would require a minimum of percolation compared to dissolution of larger meteoritic minerals; consequently, other trace elements, if present homogeneously in the larger crystal lattices, would be comparatively unaffected, because larger quantities of water would be required to dissolve these phases.

The percolation mechanism given above may, however, be invalid for the following reasons. Dufresne and Anders (1962) have noted that the magnesium abundances in all carbonaceous chondrites are relatively constant and that large quantities of magnesium sulfate have been synthesized on the spot and not transported great distances within Type I carbonaceous chondrites. A percolation mechanism most certainly would have removed appreciable fractions of the very soluble magnesium sulfate while transporting dissolved minerals containing the REE. No such transport of magnesium sulfate has been observed. On the other hand, small REE minerals could be dissolved and transported in quantities of water that would also move comparatively small amounts of the magnesium sulfate. Determination of other trace lithophilic elements having dissimilar chemical properties compared with the REE may help clarify this problem.

The analysed samples of Orgueil and Ivuna were only 1.7 and 1.0 g, respectively. However, in all other small quantities of carbonaceous and ordinary chondrites analysed by other investigators, the abundances of selected REE agreed within experimental error with those of this work and REE I.

Another serious difficulty with a percolation mechanism is that the high potential that is required for $Ce^{+3} - Ce^{+4}$ oxidation is not consistent with the presence of reduced species (e.g. FeS and hydrocarbons) in Type I carbonaceous chondrites.

In summary, the conditions for the explanation of the low $La/10^6$ Si ratio of 0.36 in Type I carbonaceous chondrites by a percolation mechanism seem to be too restrictive. In view of the absence of magnesium sulfate transport and of the high potential of Ce oxidation, the low REE abundance with respect to silicon and the different Ce/La ratios in Type I carbonaceous chondrites indicate depletion of the

whole REE series, and of Ce with respect to the other trivalent REE. This most probably occurred before the accretion of the carbonaceous chondrites in a hot portion of the solar nebula (Wood, 1962).

Reed et al. (1960), found by neutron-activation analysis a lower uranium concentration of 0·008 ppm in Orgueil compared with 0·016 ppm in Mighei, and Goles and Anders (1962) found 0·015 ppm and 0·020 ppm in Mighei and Murray, respectively. If the percolation mechanism is correct, uranium must be relegated to small soluble phases in Orgueil. Abundances of barium, another trace lithophilic and nonvolatile element, in Orgueil were similar to Mighei at 2·4 and 2·5 ppm, respectively (Reed et al., 1960). On the other hand, Pinson et al. (1953), using spectrochemical techniques, reported <1, <1, <1, and 4 ppm for Ba, Sr, Zr, and Sc abundances in Orgueil, respectively. The Sc content of 4 p.p.m. in Orgueil agrees within ∼30 per cent with the Sc value of 5·3 p.p.m. reported in this work. The hiatus between the neutron activation and spectrochemical data for the trace elements Ba, Sr, and Zr allows no clear comparisons between this data and the REE, Y, and Sc data in Orgueil.

An examination of all the Ce/La ratios in REE-I and Table 3 of this paper reveals essentially 3 arbitrary groups of Ce/La values with no clear distinction among the 5 chondritic categories. Ce/La ratios of 2·30, 2·52, 2·55, 2·69, 2·64, 2·81, and 2·81 (2·30–2·81 group) were obtained for 2 carbonaceous pigeonitic, 2 Type II carbonaceous, 2 enstatitic, and 1 hypersthenic chondrite, respectively; Ce/La ratios of 3·08, 3·50, 3·22, 3·04, 3·06 (3·04–3·50 group) exist for 2 Type I carbonaceous, 1 enstatitic, 1 bronzitic, and 1 hypersthenic chondrite, respectively. Finally, the 2 hypersthenic chondrites Holbrook and Modoc, which constitute the last group, have especially high Ce/La ratios at 4·71 and 4·40, respectively. Unreliable ratios of Ce/La for Allegan and Richardton have been neglected (these two meteorites were the first analysed). Such an apparent random distribution in Ce/La ratios complicates interpretation of the physiochemical histories of the individual meteorites.

3. Absolute and relative abundances of the REE, Y, and Sc in Mighei and Modoc agree with the average values in 17 chondrites. The single exception, Ce in Modoc, has been noted above.

It is noted by Zaozerskii and Patkin (1959) that Ce may be easily oxidized to the tetravalent state by atmospheric oxygen under increased pH and temperature conditions. If the REE are introduced into the crystallizing matrix of the chondrite, with enhancement of the Ce^{+4} abundance, it is conceivable that the abundances of other tetravalent ions, such as Ti, Zr, Hf, and Th, may be increased in these hypersthenic chondrites. Also, oxidation of the two hypersthenic chondrites Modoc and Holbrook is not inconsistent with the observation that the FeO content in hypersthenic chondrites is considerably larger (because of more oxidation) than in bronzitic chondrites.

Absolute values in Modoc of 0·080 ± 0·001 and 9·2 ppm for Eu and Sc, respectively, as determined by Bate et al. (1960), by neutron activation analysis, agree with the Eu and Sc values of this work—0·080 ± 0·004 and 8·1 ± 0·2 ppm, respectively. An estimated 3 per cent error on the Sc value of 9·2 by Bate et al., brings the Sc agreement within the 95 per cent confidence level. Such close agreement again underscores the homogeneity of chondritic meteorites.

Abundances of REE, Y and Sc in amphoteric chondrite Manbhoom

Absolute and relative REE, Y and Sc abundances in the amphoteric chondrite Manbhoom agree well with those found in the ordinary chondrites. KVASHA (1958) and MASON (1962a) have categorized this chondrite, which contains very little free metal, as an olivine hypersthene. Furthermore, the abundance in Manbhoom of another critical trace element, Cd, which is a chalcophilic element, agrees with Cd abundances of hypersthenic and bronzitic chondrites (SCHMITT *et al.*, 1963b). With a "normal" Ce/La ratio of 2·81, the hypersthenic chondrite Manbhoom is, with regard to oxidation history, more closely related to Kyushu than to Modoc and Holbrook.

Abundances of selected REE in troilite phase

Absolute concentrations of the five REE Pr, Sm, Ho, Yb and Lu in the troilite (FeS) phase of the bronzitic chondrite Richardton are ~1/3 to 1/2 as large as they are in the whole meteorite. The concentration of La at 0·74 ppm seems to be twice as large as the average La value in chondritic matter and therefore may be due to contamination. In view of the lithophilic character of the REE, such a depletion of 2 or more was anticipated in the troilite phase.

Yttrium abundances in chondrites

Previous work (REE-I) reported the absolute abundances of Y in seven chondrites to be from 1·02 to 2·42 ppm, averaging 1·9 ± 0·4 ppm, with a $Y/10^6$ Si atomic ratio of 3·6 ± 0·7. Including the five chondritic Y values reported in this work, the average Y abundance in 12 chondrites is 1·8 ± 0·4 ppm, which corresponds to 3·4 ± 0·7 $Y/10^6$ Si. This value is much lower than the SUESS–UREY (1956) atomic value of 8·9 $Y/10^6$ Si.

Again for the element Y, the atomic abundance (based on more meteorites than given in REE-I) expressed in $Y/10^6$ Si decreases from the carbonaceous to the enstatitic meteorites: 4·7 ± 0·4 in 2 Type I carbonaceous: 4·1 in 1 Type II carbonaceous; 4·8 ± 0·1 in 2 Type III carbonaceous pigeonitic; 3·4 ± 0·2 in 4 bronzitic and hypersthenic; 2·3 ± 0·5 in 2 enstatitic chondrites. Clearly, there is no appreciable fractionation of Y with respect to Si among the carbonaceous chondrites as has been observed for the REE among the carbonaceous chondrites (see above, "General Abundances in Chondritic Meteorites," observation 1), where $La/10^6$ Si = 0·36 ± 0·01 in 2 Type I carbonaceous, 0·53 ±0·07 in Type II carbonaceous, and 0·51 ± 0·08 in 2 carbonaceous pigeonitic chondrites. It seems highly improbable that any systematic error could have been introduced in the REE and Y analyses to give this striking fractionation difference between the REE and Y in the carbonaceous chondrites. Chronologically, the analysis of Orgueil followed that of the amphoteric chondrite Manbhoom. Six other meteoritic and terrestrial specimens were analysed after Orgueil and before the Ivuna analysis; these six—in chronological order—were the eclogite, Juvinas, Lafayette, Stannern, Columbia basalt, and Veramin. Since the geochemical properties of Y are very similar to those of the heavy REE (the ionic radius of Y is identical to that of Dy), it was predicted that the Y/REE or Y/La ratio should remain essentially constant for the various chondritic classes. In general, this

has been confirmed, with Y/La ratios 7·7 for Type II carbonaceous, 9·4 for carbonaceous pigeonitic, 9·0 for bronzitic and hypersthenic, and 9·6 for enstatitic chondrites. The ratio of 7·7 is based on only one Y value. However, within the Type I carbonaceous class, where Y/La = 13·1, fractionation of ~50 per cent has occurred between Y and the REE.

GREENLAND (1963) has reported fractionation of Zn and Ge within the carbonaceous chondritic group. The chalcophilic properties responsible for the Zn and Ge fractionation, may hardly be attributed to the REE and Y. Invoking a percolation mechanism for the depletion of the REE would also fractionate Y to the same degree as the heavy REE (there was no fractionation within the REE group as a whole, Ce excepted), since the solubility products of $Y(OH)_3$ and the heavy $REE(OH)_3$ are of the same magnitude. No simple differentiation mechanism is known to the authors for separating Y from the REE by ~50 per cent without associated internal REE fractionation. In fact all observed fractionations of the REE and Y in achondrites reported in this work and in REE-I have differentiated Y and Dy (same ionic radii) to practically the same degree relative to Y and Dy in all chondritic classes except the Type I carbonaceous. The absence of a plausible geochemical explanation for the REE fractionation in the carbonaceous chondrites suggests that the Type I carbonaceous chondrites may possibly have originated from a different nucleosynthetic source than the other chondrites. Changes in normalized REE abundances and in sensitive isotopic ratios serve as checks, since Y and the light REE are synthesized (BURBIDGE et al., 1957 and CLAYTON et al., 1961) predominantly by the s-process and the heavy REE principally by the r-process. With the exception of Ce, most of the REE abundances and isotopic ratios agree within experimental error, whereas the elemental abundance of Yb in Orgueil is high and unexplainable at the moment, possibly in experimental error.

Since Yb^{168} is produced via the p-process and Yb^{174} via the s- and r-processes, the agreement within the 95 per cent confidence level of the ratio Yb^{168}/Yb^{174} in Orgueil and Ivuna would rule out appreciable synthesis from another source. Also, since Yb^{168} and Yb^{174} have thermal-neutron cross sections of 11,000 and 60 barns, respectively, the 95 per cent confidence-level agreement between Yb^{168}/Yb^{174} in Type I meteoritic and terrestrial matter rules out any significant differences in thermal-neutron irradiation exposures that may have occurred for meteoritic and terrestrial matter (FOWLER et al., 1962). In addition, the near identity of the Ce^{140}/Ce^{142} isotopic ratios in Orgueil and Ivuna (see Table 1) to corresponding ratios in other meteoritic and terrestrial matter does not indicate different nucleosynthetic sources. Ce^{140} and Ce^{142} are synthesized via the s- and r-processes, respectively (BURBIDGE et al., 1957 and CLAYTON et al., 1961).

For four meteorites Forksville, Holbrook, Pasamonte and Murray (a Type II carbonaceous chondrite), MURTHY and SCHMITT (1963) have found indistinguishable isotopic ratios to ±0·5 per cent for the sensitive REE, Sm, Eu and Gd (some of whose isotopes have extremely high thermal-neutron cross sections) compared to standard terrestrial matter. Their technique, a combination of neutron-activation and mass-spectrometric methods, could check the possibility that Type I carbonaceous chondrites originated from a different nucleosynthetic source or were exposed to different thermal-neutron exposures than were other chondrites and terrestrial matter.

Abundances of REE, Y and Sc in Ca-rich achondrites

Absolute abundances of the REE and Sc in the eucritic Ca-rich achondrites Juvinas and Stannern (La = 2·53 ppm and 4·89 ppm, respectively) agree well with two previous analyses of Ca-rich achondrites, eucritic Pasamonte (La = 3·21 ppm) and howarditic Nuevo Laredo (La = 4·03 ppm) (see REE-I). More significant is the observation that normalized abundances of the REE in all four of these Ca-rich achondrites (and also Y in two achondrites) are identical to the REE in chondrites. On the average, the absolute REE and Y abundances in Ca-rich
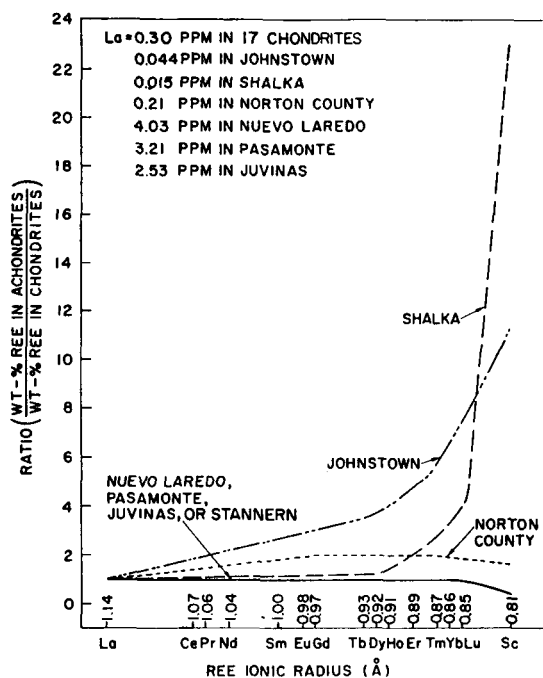
Fig. 1. Ratios of normalized wt.% of the REE in the "chondrite-like," or Ca-poor, achondrites Johnstown, Shalka, and Norton County and in the "basalt-type," or Ca-rich, achondrites Nuevo Laredo, Pasamonte, Juvinas, and Stannern to those in 17 chondrites as a function of trivalent ionic radius.

achondrites are ∼12 times more abundant in chondrites, and the Sc has been enriched ∼4 times in the four Ca-rich achondrites relative to the Sc in chondrites. If these achondrites have been derived from basic chondritic matter, an equivalent observation is that the chondritic matter has retained ∼3 times more Sc compared to its retention of the REE and Y.

A plot (Fig. 1) of the ratios of the normalized weight per cent of the REE and Y in achondrites to the normalized REE and Y in chondrites clearly shows nonfractionation of the REE and Y in Ca-rich achondrites compared to Ca-poor achondrites. However, Sc has been fractionated in Ca-poor achondrites to significant degrees (Norton County excepted) compared to the chondritic values. See REE-I for a suggested explanation based on the geochemical similarity between scandium, magnesium and divalent iron (Goldschmidt, 1954).

Fractionation of REE, Y and Sc in nakhlitic meteorites

Differences in the abundances of the REE, Y and Sc in the two nakhlitic meteorites relative to their abundances in ordinary chondrites have been recently reported by SCHMITT and SMITH (1963). In short, the similarity in REE and Y fractionation in these nakhlitic meteorites relative to the REE and Y in terrestrial basalts (see Fig. 2) suggested that these two meteorites originated from terrestrial-like volcanic action on the parent meteoritic body.
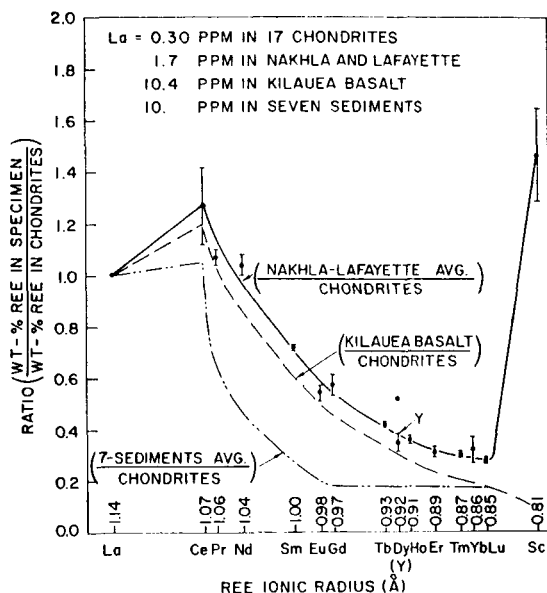
Fig. 2. Ratios of the average normalized wt.% of the REE in Nakhla and Lafayette to those in 17 chondrites: mean deviations are indicated for each REE except Dy (determined only in Lafayette). Comparative curves of ratios for REE in Kilauea Iki-22 basalt by SCHMITT *et al.* (1963a), and for the REE in average of 7 sediments by HASKIN and GEHL (1962) are included. Ratios are plotted as a function of trivalent ionic radius.

Fractionation of REE, Y and Sc in mesosideritic meteorites

Uniform fractionation (see Fig. 3) of the REE and Y relative to chondrites has been found in the silicate phase in Veramin, a mesosiderite, and an observed "fall". This contrasts sharply with nonfractionation of the REE and Y in the mesosiderite Estherville (see REE-I). On an absolute basis, the REE and Y were ~4 times more abundant in Estherville compared to chondrites vs. approximately the same REE and Y abundances in Veramin compared with chondrites. Note in Fig. 3 that Lu and Yb, the heaviest REE, are enriched by ~1·4. In the derivation of mesosiderites from some common siliceous matter, Sc has apparently been enriched to about the same degree (to a factor of ~2) in both Estherville and Veramin, since the Sc abundance in these two mesosiderites is 17·8 ppm and 16·2 ppm, respectively.

The element Eu has been enriched in Veramin by ~1·4 relative to Eu in chondrites (see Fig. 3). This is the first meteorite among 32 analysed in which Eu has been

enriched. Only in the Ca-poor achondrites and pallasites has Eu been depleted, i.e. in Norton County by ~4·2, in Johnstown by ~3·3, and in Brenham by ~1·8 (see below). This suggests that some of the mesosiderites may be the complements of Ca-poor achondrites and pallasites. The availability of a comparatively stable divalent oxidation state for Eu relative to the adjacent medium weight REE in the
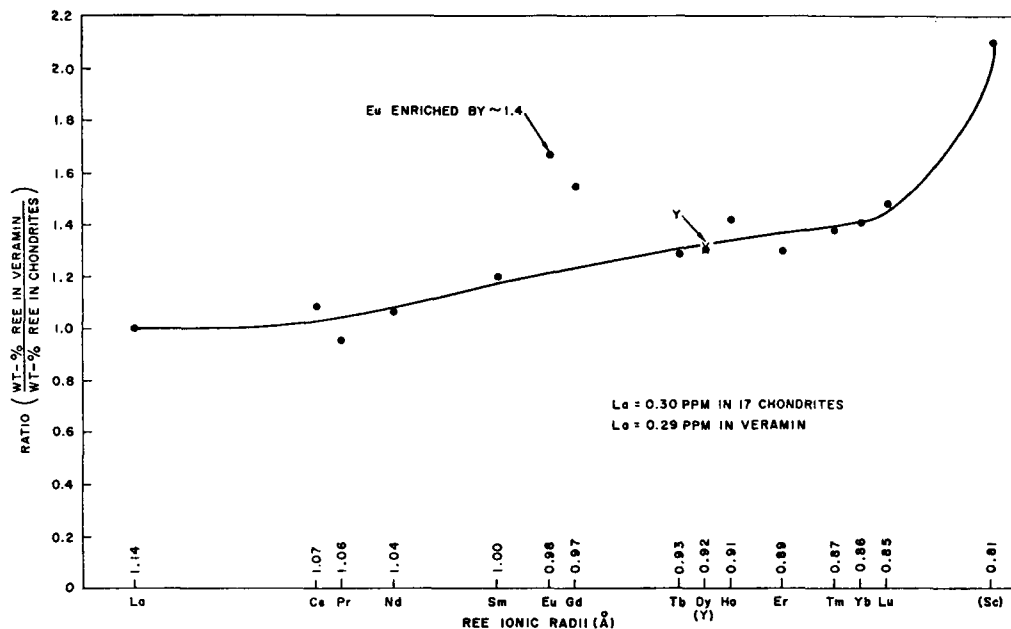

Fig. 3. Ratios of normalized wt.% of REE, Y and Sc in the mesosiderite Veramin to those in 17 chondrites as a function of trivalent ionic radius.

trivalent state may readily account for the enchancement and complementary depletion of the Eu during differentiation under appropriate reduction conditions (see REE-I).

No great confidence should be placed on the apparent Gd enrichment of ~1·24 in Veramin relative to chondrites. Because of possible overlapping of Eu and Gd during the ion-exchange elution, the Gd abundance may actually be lower than reported.

Fractionation of REE, Y and Sc in pallasitic meteorites

Absolute abundances of the REE, Y and Sc in the olivine phase of the two pallasites Brenham and Thiel Mountains were about 0·05 to 0·5 times the abundances in chondritic meteorites. In both pallasites (both were "finds"), the REE, Y and Sc have been fractioned differently (see Figs. 4 and 5). The question of terrestrial contamination may not be entirely ruled out. E. P. Henderson selected olivine crystals with the least (if any) contamination. Fractionation of the REE in Thiel Mountains strongly resembles terrestrial fractionation of REE found in Kilauea Iki-22 basalt and peridotite and in seven terrestrial sediments by Haskin and Gehl (1962) (see REE-I); therefore, terrestrial contamination of the Thiel Mountains may not be

completely ruled out. In Brenham, the fractionation of the REE, Y and Sc is completely different from terrestrial basalts, peridotites etc. Certain rare-earth minerals, such as samarskite [(Gd, U, Fe)(Nb, Ta,)$_2$O$_6$], enrich the medium-weight REE by factors of ~100 to ~40 with respect to La and Lu, respectively (SEMENOV and BARINSKII, 1958). Enrichment factors have been calculated relative to normalized
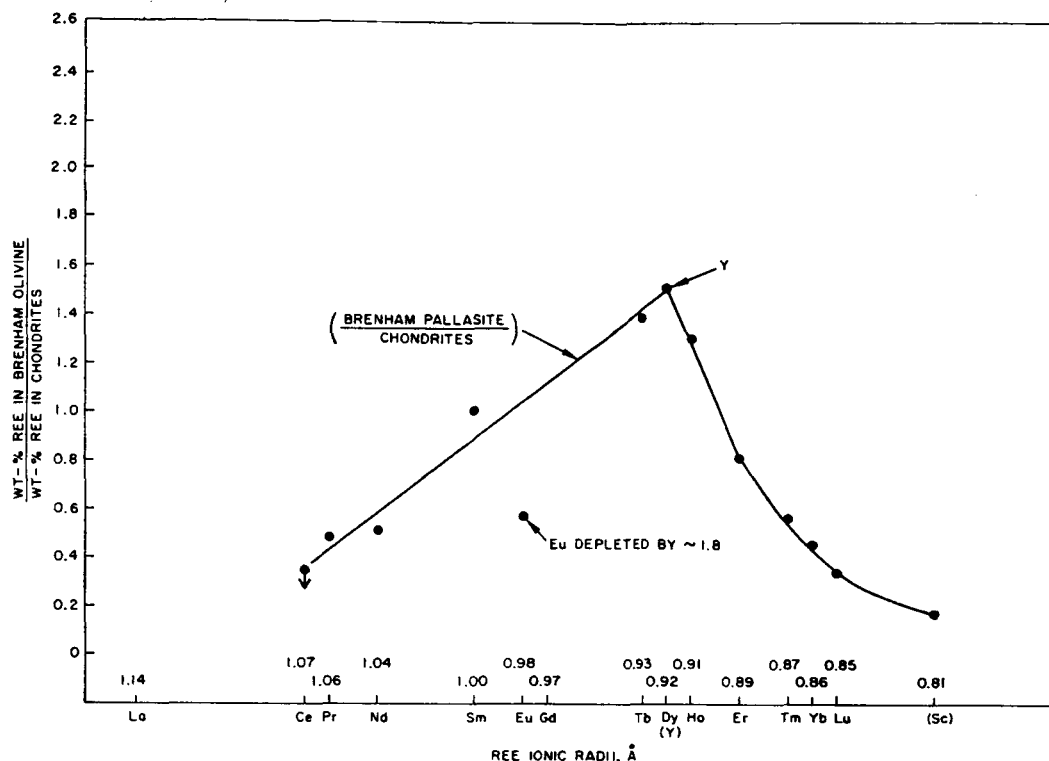

Fig. 4. Ratios of the normalized (to Sm at 0·66) wt.% REE in Brenham pallasite (olivine phase) to the average normalized wt.% REE in 17 chondrites as a function of ionic radii in Å units of trivalent REE.

REE abundances in chondritic meteorites calculated by CORYELL *et al.* (1962). In samarskite the element Eu was depleted by a factor of ~30 compared to a factor of ~1·8 in the olivine phase in Brenham. The terrestrial REE distribution of a basalt in combination with a samarskite mineral might conceivably show a composite REE distribution as observed in Brenham. With a reduced ionic radius of 0·89 Å (AHRENS, 1952) for Eu^{+2}, some Eu^{+2} might be retained by diadochic replacement in the ferromagnesian olivine phase.

In a discussion of the differentiation of chondrites and the formation of irons, stony irons and achondrites, RINGWOOD (1961) suggests that five parts of pallasites are complementary to one part of eucrite, with mesoiderites resulting from mixing or incomplete differentiation from eucrite, pallasitic and parental chondritic matter. In short, conservation of REE, Y and Sc abundance in these four different meteorite types does not adequately support the Ringwood suggestion; see SCHMITT (1962) for details.
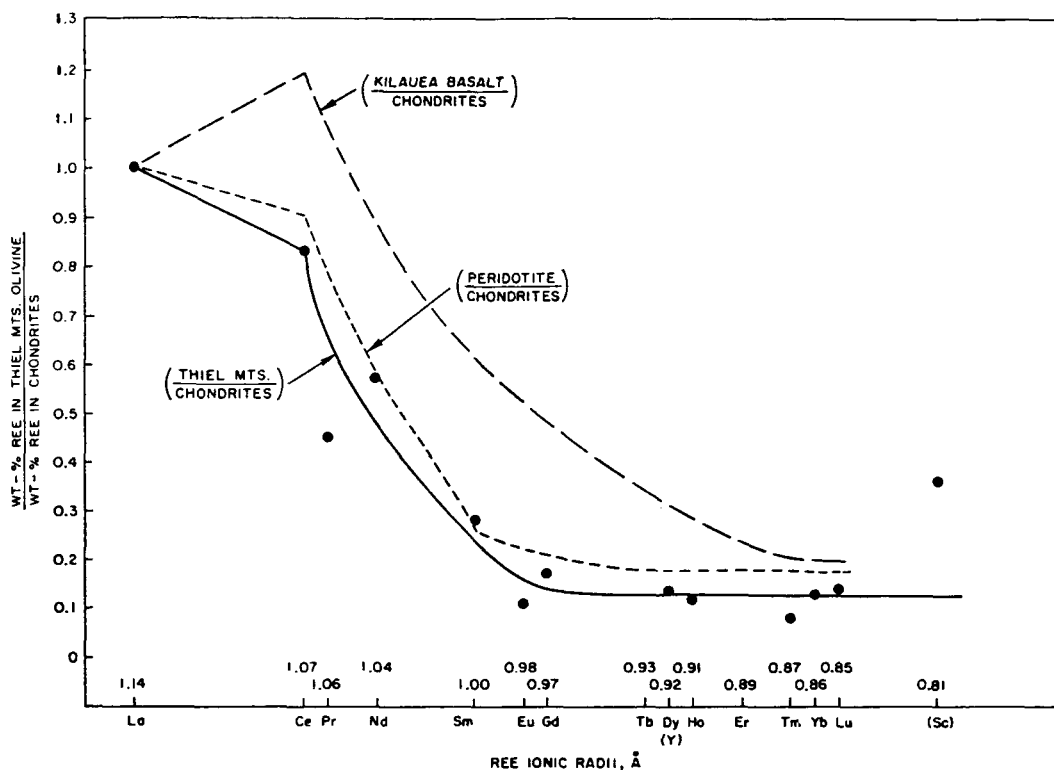
Fig. 5. Ratios of the normalized wt.% in REE in Thiel Mountains pallasite (olivine phase) to the average normalized wt.% REE in 17 chondrites as a function of ionic radii in Å units of trivalent REE. For comparison, the corresponding ratios for terrestrial Kilauea Iki-22 basalt and Wesselton Mine peridodite are included (see REE-I).

Fractionation of REE, Y and Sc in terrestrial matter

Relative to the REE, Y and Sc in chondrites, fractionation of these elements has occurred (see Fig. 6) in a hornblende eclogitic inclusion from a basic breccia pipe near Delegate, N.S.W., Australia. Lovering (1962) states that this specimen is an unaltered sample of basic upper-mantle material, and that the Rb–Sr data suggests an age of $4 \cdot 3 \pm 0 \cdot 2 \times 10^9$ years for it. The absolute abundances of the REE contents are roughly similar (see Table 4 and Fig. 6) for La, Lu and Sc in the South African and Australian eclogite. According to Lovering (1962) the trace-element contents of the South African eclogite may be dubious because of possible contamination by basalt. However, from a consideration of the absolute abundances and distributions of the REE in oceanic basalts (see dashed curve of Fig. 2), it seems improbable that the African eclogite has been seriously contaminated by REE from basalts. Also, note that Ce has differentiated as expected, which would indicate that there was no special oxidation history for these eclogites compared with chondrites.

The REE distribution for the earth's crust may be best represented by that found in 7 different sediments (see Haskin and Gehl, 1962 and Schmitt *et al.*, 1963c). Assuming an initial chondritic distribution for the earth for the non-volatile elements

Table 4. Absolute and normalized abundances of REE, Y and Sc in an Australian eclogite and tholeiitic basalt (Columbia Plateau) as determined by neutron-activation analysis

Isotope measured	Radionuclide	Absolute abundances in 10^{-6} g/g sample (ppm)		Normalized abundances	
		Eclogite	Basalt	Eclogite	Basalt
La^{139}	40-h La^{140}	3·57 ± 0·07	25·3 ± 0·5	1·00	1·00
Ce^{140}	32-d Ce^{141}	14·8 ± 0·2	64·0 ± 1·0	3·95	2·60
Ce^{142}	33-h Ce^{143}	13·4 ± 0·3	67·3 ± 1·4		
Pr^{141}	19-h Pr^{142}	2·51 ± 0·06	7·90 ± 0·16	0·70	0·31
Nd^{146}	11·1-d Nd^{147}	11·7 ± 0·2	36·6 ± 0·6	3·28	1·45
Sm^{152}	47-h Sm^{153}	3·84 ± 0·03	10·0 ± 0·2	1·08	0·40
Eu^{151}	9·3-h Eu^{152m}	1·24 ± 0·02	2·32 ± 0·03	0·35	0·092
Eu^{151}	13-y Eu^{152}	1·26 ± 0·04	2·28 ± 0·05		
Gd^{158}	18-h Gd^{159}	5·17 ± 0·09	10·2 ± 0·2	1·45	0·40
Tb^{159}	72-d Tb^{160}	0·75 ± 0·01	1·66 ± 0·02	0·21	0·066
Dy^{164}	2·3-h Dy^{165}	5·7 ± 0·5	9·6 ± 0·2	1·6	0·38
Ho^{165}	27-h Ho^{166}	1·00 ± 0·01	1·94 ± 0·01	0·28	0·077
Er^{168}	9·4-d Er^{169}	2·62 ± 0·10	5·60 ± 0·24	0·77	0·214
Er^{170}	7·5-h Er^{171}	2·88 ± 0·03	5·25 ± 0·08		
Tm^{169}	129-d Tm^{170}	0·41 ± 0·02	0·75 ± 0·02	0·115	0·030
Yb^{168}	32-d Yb^{169}	1·95 ± 0·06		0·57	
Yb^{174}	4·2-d Yb^{175}	2·05 ± 0·03	3·88 ± 0·06		
Lu^{167}	6·8-d Lu^{177}	0·32 ± 0·01	0·64 ± 0·01	0·088	0·0253
Y^{89}	64-h Y^{90}	27 ± 0·4	53 ± 3·0	7·6	2·08
Sc^{45}	85-d Sc^{46}	48 ± 1	37 ± 0·8	13·6	1·46

and also assuming that there was no appreciable mixing in the mantle, the REE distribution in the upper mantle will be uniformly enriched in the heavy REE by a factor of ~5 and depleted monotonically in the light REE. Neither of the eclogitic specimens represent such a distribution. Within the framework of the assumptions given above, neither of these two eclogitic specimens represent unfractionated upper-mantle matter.

Fractionation (Fig. 7) of the REE, Y and Sc in the Columbia Plateau basalt appears to be similar to that found in Kilauea Iki-22 basalt (see REE-I). The severity of differentiation for the REE and Y in the Kilauea basalt exceeds that in the Columbia Plateau basalt, whereas nearly the same degree of differentiation has occurred in these basalts for Sc. If the curves from Gd to Ce are extrapolated back
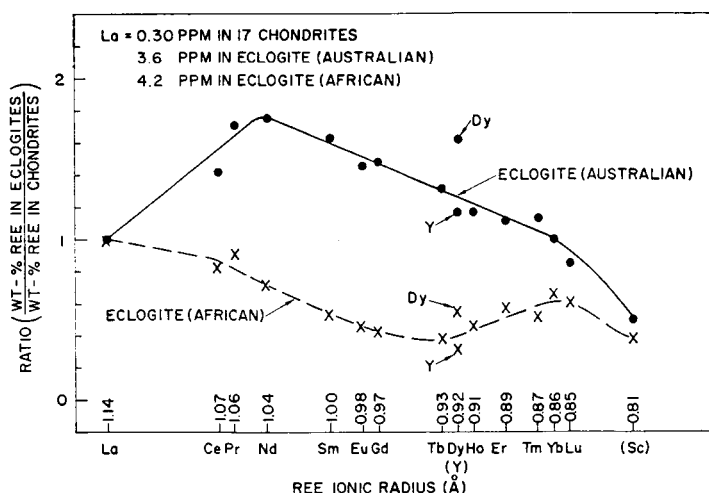
Fig. 6. Ratios of the normalized wt.% of the REE in an Australian eclogite and South African eclogite to those in 17 chondrites as a function of trivalent ionic radius.
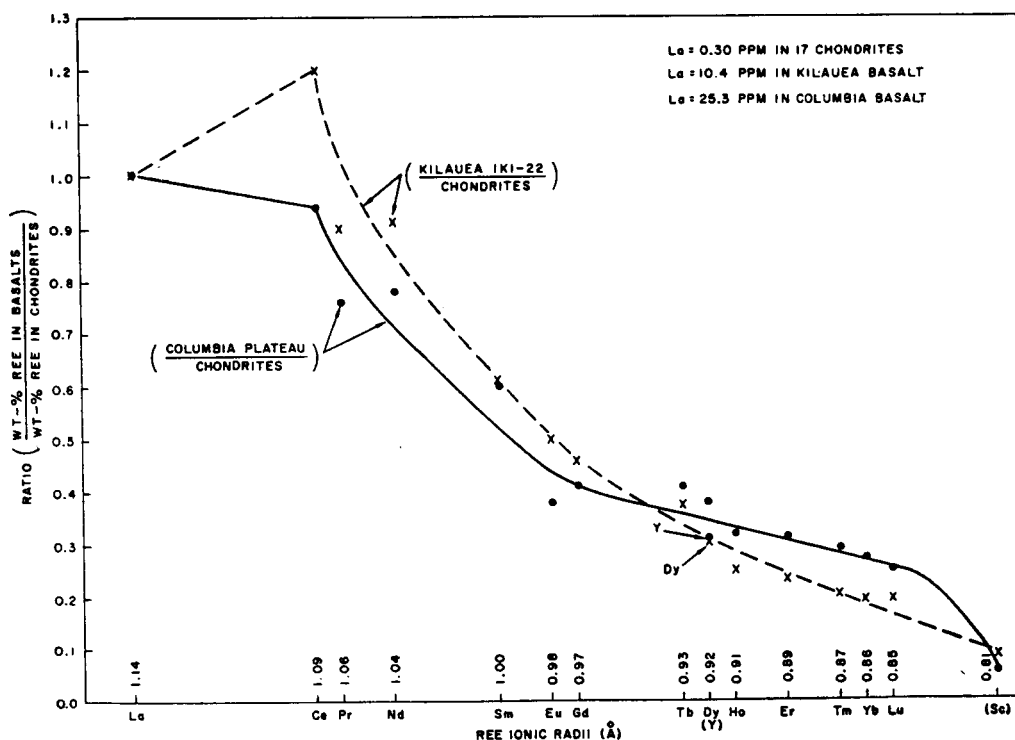

Fig. 7. Ratios of normalized wt.% of the REE, Y and Sc in Columbia Plateau tholeiitic basalt and in Kilauea Iki-22 basalt to those in 17 chondrites as a function of trivalent ionic radius.

to La, it appears that La has been fractionated more in Kilauea than in Columbia plateau basalt. The general similarity in REE and Y fractionation in two basalts compared to that observed in two nakhlitic achondritic meteorites (see Fig. 2) strongly suggests that the nakhlites originated from terrestrial-like volcanic action.

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